

WE CLAIM:

1. An advanced sealing (gasket) member for sealing in high temperature devices, comprising:
a mica based member wherein said member comprises an infiltrating material whereby low effective leak rates are obtained.
2. The sealing member of claim 1 wherein said mica based member comprises a mica selected from the group consisting of Phlogopite, Muscovite, Biotite, Fuchsite, Lepidolite, Zinnwaldite, paper, flakes, filaments, fragments, particles, and combinations thereof.
3. The sealing member of claim 1 wherein said infiltrating material is a member selected from the group consisting of H_3BO_3 , $\text{Bi}(\text{NO}_3)_3$, B_2O_3 , Bi_2O_3 , glass former, melt former, glass, oxides, mica:glass composite(s), or combinations thereof.
4. The sealing member of claim 3 wherein said glass is G-18 glass.
5. The sealing member of claim 1 wherein leak rates are less than $1\text{E-}02$ sccm/cm at operating temperatures in the range from 600°C to about 850°C .
6. The sealing member of claim 1 wherein said member comprises a mica:glass composite having a mica-based concentration up to about 50% by volume.
7. The sealing member of claim 1 wherein said sealing member comprises a mica:glass composite having 90% by volume of a mica based material and 10% by volume of a glass forming material.

8. The sealing member of claim 1 wherein said sealing member comprises a mica:glass composite mixture of 80% by volume of a mica-based material and 20% by volume of a glass forming material.

9. The sealing member of claim 1 wherein said infiltrating material comprises a G-18 glass.

10. A multilayer (compressive) seal having superior thermal cycling stability for sealing in high temperature devices, the seal comprising:

a sealing (gasket) member defining first and second opposing surfaces;
a first compliant interlayer disposed adjacent with said first surface;
a second compliant interlayer disposed adjacent said second surface;

and,

wherein said sealing member is infiltrated with at least one glass or melt forming material whereby a plurality of spaces within said member are effectively sealed and low effective leak rates are achieved.

11. The seal according to claim 10, wherein leak rates are less than $1\text{E-}02$ sccm/cm at operating temperatures in the range from 600°C to about 850°C .

12. The seal in accordance with claim 10 wherein the sealing (gasket) member comprises a mica selected from the group consisting of Phlogopite, Muscovite, Biotite, Fuchsite, Lepidolite and Zinnwaldite, paper, flakes, filaments, fragments, particles, and combinations thereof.

13. The seal in accordance with claim 10 wherein at least one of said first and second compliant interlayers comprises a member selected from the group consisting of a glass, a glass-ceramic, a mica glass-ceramic, a phase-

separated glass, a glass composite, a cermet, a metal, a metal foil, a metal alloy, a metal composite, a mica-glass composite, or combinations thereof.

14. The seal in accordance with claim 10 wherein said sealing member comprises a mica:glass composite having a mica-based concentration up to about 50% by volume.

15. The seal in accordance with claim 10 wherein said sealing member comprises a mica:glass composite having 90% by volume of a mica based material and 10% by volume of a glass forming material.

16. The seal in accordance with claim 10 wherein said sealing member comprises a mica:glass composite mixture of 80% by volume of a mica-based material and 20% by volume of a glass forming material.

17. The seal in accordance with claim 15 wherein said glass forming material comprises a G-18 glass.

18. The seal in accordance with claim 16 wherein said glass forming material comprises a G-18 glass.

19. A process for making a sealing (gasket) member exhibiting superior thermal cycle stability and low effective leak rates, the steps comprising:

providing an infiltrating liquid at a first temperature;
infiltrating a plurality of spaces in a sealing (gasket) member with said liquid; and
fixing said material at a second temperature within said member whereby low effective leak rates are obtained.

20. The process in accordance with claim 19 wherein said spaces comprise a member selected from the group consisting of voids, spaces, flow paths, leak paths, gaseous flow paths, continuities, necking areas, interstitial spaces, three-dimensional spaces, and combinations thereof.

21. The process in accordance with claim 19 wherein said infiltrating liquid comprises at least one material selected from the group consisting of H_3BO_3 , $\text{Bi}(\text{NO}_3)_3$, B_2O_3 , Bi_2O_3 , glass former, melt former, glass, mica:glass composite(s), or combinations thereof.

22. The process in accordance with claim 19 wherein said liquid further comprises materials having a melting point below about 850 °C.

23. The process in accordance with claim 19 wherein said member comprises a mica selected from the group consisting of Phlogopite, Muscovite, Biotite, Fuchsite, Lepidolite and Zinnwaldite, paper, flakes, filaments, fragments, particles, and combinations thereof.

24. The process in accordance with claim 19 wherein said first temperature is in the range from 1° C to about 90° C.

25. The process in accordance with claim 19 wherein said second temperature is in the range from about 50 °C to 150 °C for a period of from 0.5 to 1 hour.

26. The process in accordance with claim 19 wherein said leak rates are less than 1E-02 sccm/cm at operating temperatures in the range from 600 °C to 850 °C.

27. A process for making a sealing (gasket) member exhibiting superior thermal cycle stability, the steps comprising:

providing an infiltration material at a first temperature;

forming a sealing (gasket) member comprising said infiltration material;

and

fixing said material in said sealing member at a second temperature

whereby a plurality of spaces within said member are made discontinuous and low effective leak rates are obtained.

28. The process of claim 27, wherein said first temperature is in the range from 1 °C to about 90 °C.

29. The process of claim 27, wherein said second temperature is in the range from about 50 °C to 150 °C for a period of from 0.5 to 1 hour.

30. The process of claim 27 wherein said leak rates are less than 1E-02 sccm/cm at operating temperatures in the range from 600 °C to 850 °C.

31. The process of claim 27 wherein said infiltration material comprises a mica:glass composite having a glass-based concentration up to about 50% by volume.

32. The process of claim 27 wherein said infiltration material comprises a mica:glass composite having 90% by volume of a mica based material and 10% by volume of a glass forming material.

33. The process of claim 27 wherein said material is a composite mixture comprising 80% by volume of a mica-based material and 20% by volume of a glass forming material.

34. The process of claim 32 wherein said mica-based material is selected from the group consisting of Phlogopite, Muscovite, Biotite, Fuchsite, Lepidolite and Zinnwaldite, paper, flakes, filaments, fragments, particles, and combinations thereof.

35. The process of claim 33 wherein said mica-based material is selected from the group consisting of Phlogopite, Muscovite, Biotite, Fuchsite, Lepidolite and Zinnwaldite, paper, flakes, filaments, fragments, particles, and combinations thereof.

36. The process of claim 27 wherein said spaces comprise a member in the group consisting of voids, flow paths, leak paths, three dimensional spaces, interstices, continuities, necking areas, and combinations thereof.

37. A process for making a multi-layer (compressive) seal having superior thermal cycle stability for high-temperature electrochemical applications and structures, comprising:

providing a sealing (gasket) member comprising an infiltration material wherein said member defines first and second generally flat opposing surfaces; and

applying a compliant material to said first and second surfaces to form first and second compliant interlayers;

38. The process in accordance with claim 37 wherein said providing comprises a material selected from the group consisting of glass forming

material(s), melt forming material(s), mica:glass composite(s), or combinations thereof.

39. The method of claim 37, wherein said member comprises at least one glass or melt forming material selected from the group consisting of H_3BO_3 , $\text{Bi}(\text{NO}_3)_3$, B_2O_3 , Bi_2O_3 , glass forming material(s), melt forming material(s), composite(s), mica:glass composite(s), and combinations thereof.

40. The method of claim 37 wherein said materials has a melting point below about 850 °C.

41. The method in accordance with claim 37 wherein said providing comprises a member selected from the group consisting of slip casting, tape casting, tape calendaring, die pressing, or combinations thereof.

42. The method in accordance with claim 37 wherein at least one of the first and second compliant interlayers comprises a member selected from the group consisting of a glass, a glass-ceramic, a mica glass-ceramic, a phase-separated glass, a glass composite, a cermet, a metal, a metal foil, a metal alloy a metal composite, or combinations thereof.

43. The method in accordance with claim 37 wherein said sealing member has a thickness of from about 25 microns to about 1 millimeter.

44. The method in accordance with claim 37 wherein each of the first and second compliant interlayers has a thickness of from about 0.005 millimeters to about 1 millimeter.

45. A method for making a multi-layer seal having superior thermal cycle stability for high-temperature electrochemical applications and structures, comprising:

providing a sealing (gasket) member wherein said member defines first and second generally flat opposing surfaces.

applying a compliant material to said first and second surfaces to form first and second compliant interlayers; and

wherein said member is infiltrated with at least one glass or melt forming material.

46. The method in accordance with claim 45 wherein said glass (or melt) forming material is selected from the group consisting of H_3BO_3 , $\text{Bi}(\text{NO}_3)_3$, B_2O_3 , Bi_2O_3 , glass, mica:glass composite(s), or combinations thereof.

47. The method of claim 45 wherein said glass forming material comprises a G-18 glass.

48. The method in accordance with claim 45 wherein said providing comprises a member selected from the group consisting of dip-coating, painting, screen printing, deposition, sputtering, tape casting and sedimentation.

49. A method for making a multi-layer (compressive) seal having superior thermal cycle stability for high-temperature electrochemical applications and structures, comprising:

providing a glass infiltrated sealing (gasket) member defining first and second generally flat opposing surfaces;

applying a compliant material to said first and second surfaces to form first and second compliant interlayers; and,

wherein said seal is under a compressive stress.

50. An electrochemical device, comprising:

a plurality of components, said components forming at least one boundary between diverse gaseous streams and defining at least one junction therebetween;

a multi-layer compressive seal positioned at the junction, the seal composed of a sealing (gasket) member comprising an infiltrating material said sealing member disposed between two compliant interlayers, wherein each compliant interlayer is disposed between the sealing member and one of said components; and

a compression member for exerting a compressive force to the components and the sealing member.

51. The electrochemical device in accordance with claim 50 wherein the sealing (gasket) member comprises an infiltrated mica.

52. The electrochemical device in accordance with claim 50 wherein at least one of said compliant interlayers comprises glass.

53. The electrochemical device in accordance with claim 50 wherein at least one of said compliant layers comprises a metal.

54. The electrochemical device in accordance with claim 50 wherein the glass has a softening point lower than or equal to the operating temperatures of the device.

55. The electrochemical device in accordance with claim 50 wherein the glass is non corrosive to surfaces of the components in contact with the glass under operating conditions.

56. The electrochemical device in accordance with claim 50 wherein the metal is resistant to oxidation under operating conditions of the device.

57. The electrochemical device in accordance with claim 50 wherein the compliant interlayer is a metallic foil having a thickness of from about 0.005 millimeters to about 1 millimeters prior to heating.

58. The electrochemical device in accordance with claim 50 wherein the compressive force is a force of from about 5 to about 500 psi.

59. The electrochemical device in accordance with claim 50 wherein said sealing (gasket) member has a thickness of from about 25 microns to about 2 millimeters.

60. The electrochemical device in accordance with claim 50 wherein each of said compliant layers has a thickness of from about 0.005 millimeters to about 1 millimeter prior to heating.

61. The electrochemical device in accordance with claim 50 wherein the electrochemical device comprises a member selected from the group consisting of a solid oxide fuel cell, a syngas membrane reactor, an oxygen generator.

62. A solid oxide fuel cell assembly for electrochemically reacting a fuel gas with a flowing oxidant gas at an elevated temperature to produce a DC output voltage, said solid oxide fuel cell comprising:

a plurality of generally planar integral fuel cell units, each unit comprising a layer of ceramic ion conducting electrolyte disposed between a conductive anode layer and a conductive cathode layer, wherein said units are arranged one on another along a longitudinal axis perpendicular to said planar units to form a fuel cell stack;

a multi-layer non-conducting seal disposed between the anode layer and the cathode layer of adjacent fuel cell units, wherein the seal is composed of an infiltrated sealing (gasket) member disposed between two compliant interlayers; and

a compression member for exerting a compressive force along the longitudinal axis.

63. The assembly in accordance with claim 62 wherein the compressive force is in the range from about 5 to about 500 psi.

64. A method for sealing a junction between adjacent ceramic or metallic components of an electrochemical device, comprising:

positioning between the adjacent components a multi-layer seal comprising a sealing member infiltrated with at least one glass forming material wherein said member is disposed between a first compliant interlayer and a second compliant interlayer, wherein each of the first and second compliant interlayers is positioned between the sealing member and one of the components; and

applying a compressive force to the components and the seal.

65. The method in accordance with claim 64 wherein the sealing member comprises a member selected from the group consisting of melt forming materials, glass forming materials, glass, mica:glass composite materials, H_3BO_3 , B_2O_3 , $\text{Bi}(\text{NO}_3)_3$, Bi_2O_3 , G-18 glass, or combinations thereof.

66. A solid oxide fuel cell assembly for electrochemically reacting a fuel gas with a flowing oxidant gas at an elevated temperature to produce a DC output voltage, said solid oxide fuel cell comprising:

a plurality of generally planar integral fuel cell units, each unit comprising a layer of ceramic ion conducting electrolyte disposed between a conductive anode layer and a conductive cathode layer, wherein said units are arranged one on another along a longitudinal axis perpendicular to said planar units to form a fuel cell stack;

a multi-layer non-conducting seal disposed between the anode layer and the cathode layer of adjacent fuel cell units, wherein the seal is composed of a sealing member infiltrated with at least one glass forming material or composite(s) disposed between two compliant interlayers; and

a compression member for exerting a compressive force along the longitudinal axis.

67. The assembly in accordance with claim 66 wherein the compressive force is a force of from about 5 to about 500 psi.

68. The assembly in accordance with claim 66 wherein the anode layer is composed of a first porous ceramic material and the cathode layer is composed of a second porous ceramic material.